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**THE "BLEACHING" EFFECT IN $ZnS(Co)$
CRYSTALS UNDER THE INFLUENCE
OF EXTREMELY LARGE PULSES
OF A RUBY LASER**

by L. N. Galkin

Doklady Akademii Nauk SSSR,
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UNDER THE INFLUENCE OF EXTREMELY LARGE PULSES
OF A RUBY LASER

By L. N. Galkin

Translation of "Effekt 'prosvetleniya' v kristallakh $\text{ZnS}(\text{Co})$
pod deystviyem gigantskikh impul'sov rubinovogo opticheskogo
kvantovogo generatora."
Doklady Adademii Nauk SSSR, Vol. 170, No. 2, pp. 315-316, 1966.

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THE "BLEACHING" EFFECT IN ZnS(Co) CRYSTALS UNDER THE INFLUENCE OF
EXTREMELY LARGE PULSES OF A RUBY LASER

(Presented by Academician A. A. Lebdev, December 8, 1965)

L. N. Galkin

The transmission spectrum of cobalt-doped ($N_{\text{Co}} = 2 \times 10^{18} \text{ cm}^{-3}$) zinc sulfide crystals (1 mm thick) under the effect of a high-intensity ruby laser radiation was investigated at 694 mμ. The experiments were carried out by means of the setup shown in Figure 1. The results indicate that 50% "bleaching" was attained at an incident intensity of approximately 50 Mw/cm², in a process during which a considerable portion of the Co^{2+} ions were excited from the $^4\text{A}_2(\text{F})$ state to the $^4\text{T}_1(\text{P})$ state. The relaxation time of the inverted transition was found to be high, $\tau = 1 \cdot 10^{-9}$ sec, assuming $\sigma = 1.6 \cdot 10^{-17} \text{ cm}^{-2}$. This indicates that only a negligible number of ions remains in the metastable $^4\text{T}_2(\text{F})$ state, transition from which into the lower $^4\text{A}_2(\text{F})$ state is radiative and requires tens of μsec. Thus, the $^4\text{T}_1(\text{P}) \rightarrow ^4\text{A}_2(\text{F})$ and $^4\text{T}_2(\text{F}) \rightarrow ^4\text{A}_2(\text{F})$ transitions remain quasi-independent when the Co ions are exposed to short optical pulses.

The ability of several substances to change their transmission under /315* the influence of powerful light fluxes has been recently discovered (Ref. 1). This article describes the effect of "bleaching" in a crystal of zinc sulfide which is activated by cobalt, when it is irradiated by a powerful ruby laser.

At the present time we may formulate certain principles which make it possible to predict whether bleaching will be observed in a given substance in the case of light fluxes whose strength may be achieved in practice**. In the first place, a substance in the ground state which absorbs light in the requisite region of the spectrum may be used as the coloration source.*** In the second place, there must be no absorption of light by a

* Numbers in the margin indicate pagination in the original foreign text.

** We shall investigate substances to which source coloration models may be applied (elements and ions with unfilled d- or f- shells, organic dyes).

*** It is sometimes possible to shift the absorption band of the source toward the requisite side by the selection of the matrix, since the distance between the energy terms of the source is determined to a significant degree by the inner force interaction between the source and the matrix.

source which is in an excited state*. In the third place, the matrix itself must also be transparent for the given region of the spectrum. Finally, in the fourth place, it is desirable that the given spectrum have photoluminescence. The latter condition determines a certain "retardation" of the transitions between the source terms. Due to this fact, it is possible to observe the radiative transitions, whose probability is, as a rule, considerably less than the non-radiative transitions.

Zinc sulfide activated by cobalt satisfies all of these conditions. It was shown in (Ref. 2) that a cobalt ion Co^{2+} has several absorption bands in the visible and infrared regions of the spectrum. For us, it is important that one of them belongs to the region of radiation of a ruby laser. Figure 1 shows the spectrum, which was recorded on a SF-4 spectrometer, for the transmission of a crystal of zinc sulfide activated by cobalt. With a cobalt concentration of $N_{\text{Co}} = 2 \cdot 10^{18} \text{ cm}^{-3}$ and a sample thickness of 1 mm its transmission at the wave length 694 mμ comprised 4%.

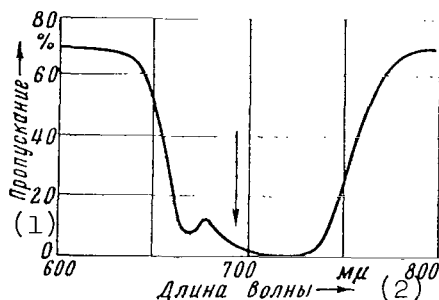


Figure 1

Transmission Spectrum of a ZnS Crystal Activated by Cobalt $N_{\text{Co}} = 2 \cdot 10^{18} \text{ cm}^{-3}$, Thickness 1 mm. The Arrow Designates the Wave Length of the Ruby laser.

(1) - Transmission; (2) - wave length.

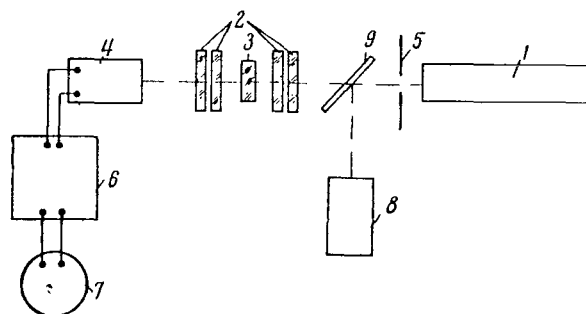


Figure 2

Diagram Showing the Arrangement for Measuring the Non-Linearity of the Transmission of the Saturable Filters.

The transmission in the case of large light fluxes was measured on a device (see Figure 2) consisting of a single pulse ruby laser 1, a system of neutral density light filters 2 with the total density $D_{10} = 2.5$, the sample to be studied 3, and the radiation receiver 4. The flux falling on the sample was changed by mutually interchanging it and the neutral/light filters. The diaphragm 5 was located on the radiation path of the laser; this diaphragm separated the portion of the flux which was the most uniform over the cross

* This does not apply to systems having a metastable state which absorb light at the working wave length [see, for example, (Ref. 3)].

section. The radiation was recorded by means of a sensitive colorimeter 4. The energy falling upon it was determined by the magnitude of the thermo-electromotive force from the thermocouples which were built in. A FEOU-15 amplifier 6 was connected between the thermocouples and the output device 7. The stability of the generator operation was controlled according to the energy falling on the auxiliary colorimeter 8. Radiation upon this colorimeter was controlled by means of a filter 9.

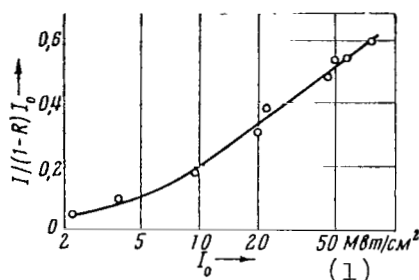


Figure 3

Transmission Change $I/(1-R)I_0$ (R - Reflection Coefficient) of a ZnS Crystal Activated by Cobalt, as a Function of the Incident Intensity I_0 of a Ruby laser.

(1) - 50 Mw/cm²

relaxation time τ is as follows, according to (Ref. 3)

$$\tau = \frac{1}{\sigma I_0} (D_e - \ln 2);$$

Assuming $\sigma = 1.6 \cdot 10^{-17}$ cm⁻², we obtain $\tau \approx 1 \cdot 10^{-9}$ seconds for our sample. For ions in an excited state (n/N_{Co}), it comprises $n/N_{Co} = (D_e - \ln 2) / [D_e - \ln 2 + 1]$ or, in our case, $n/N_{Co} \approx 0.42$.

The rapid relaxation between the states $^4T_1(P)$ and $^4A_2(F)$ indicates that only an insignificant portion of the ions is in the metastable state $^4T_2(F)$. The transition from this state into the lower state $^4A_2(F)$ has a relaxation time of tens of microseconds (Ref. 4) and is accompanied by radiation. Consequently, the $^4T_1(P) \rightarrow ^4A_2(F)$ and $^4T_2(F) \rightarrow ^4A_2(F)$ transitions are quasi-independent when ions are irradiated by short light impulses.

Figure 3 presents the results /316 derived from measuring the bleaching of a filter of ZnS(Co) as a function of the strength of the incident radiation I_0 . A bleaching of 50% was achieved for a strength of the incident flux of $I_0 \approx 50$ Mw/cm².

During the bleaching process, a significant portion of the cobalt ions Co^{2+} changed from the lower state $^4A_2(F)$ into the state $^4T_1(P)$. Let us determine the relaxation time of the inverse transition. For this purpose, we must assume that the form of the single pulse of the laser is rectangular, and its duration is such that stationary equilibrium is established in the system. For this case (bleaching equalling 50%), the constant of the

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